

HASHIBA

## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the approach of sterilizing the inside of a closed space by formaldehyde gas.

[0002]

[Description of the Prior Art] As the approach of the environmental sterilization in bioclean rooms, such as a \*\*\*\*\* room and a hospital operating room, using formalin gas is performed from the former.

[0003] However, the starting formalin gas has many which usually carry out generating supply by suitable carrier gas under heating of formalin aqueous solution. Under the present circumstances, sufficient thing to do for time amount generating maintenance mainly becomes difficult from control of formalin gas being difficult about the formaldehyde of sufficient concentration. Therefore, it was difficult to demonstrate a desirable bactericidal effect with sufficient repeatability. Furthermore, there was also a problem that a paraformaldehyde and a formic acid carried out residual contamination after sterilization processing.

[0004]

[Problem(s) to be Solved by the Invention] This invention aims at offering the sterilization approach of a closed space with little residual contamination by formaldehyde gas.

[0005]

[Means for Solving the Problem] this invention person solved the trouble of the sterilization approach by conventional formalin, and repeatability was good and he completed [ the formaldehyde gas of sufficient concentration ] a header and this invention for the sufficient approach of carrying out time amount generating maintenance.

[0006] namely, the approach this invention sterilizes the inside of a closed space -- it is -- the temperature in this closed space -- within the limits of 20-40 degrees C -- and the approach characterized by maintaining formaldehyde in concentration of at least 2000 ppm or more is offered in this closed space, maintaining the humidity in this closed space in the range of 50% - 90% of relative humidity.

[0007] Furthermore, this invention is the approach of the above-mentioned publication, and offers the approach characterized by generating said formaldehyde from a methanol or a paraformaldehyde.

[0008] The approach concerning this invention of the above-mentioned publication shows the sterilization effectiveness in a biological indicator (BI), and (ISO standard: B.Stearo:106, B.Subt:106) as a bactericidal effect.

[0009] Hereafter, it is based on the gestalt of operation and this invention is explained to a detail.

[0010]

[Embodiment of the Invention] The outline of a gestalt of enforcing the approach concerning this invention is shown in drawing 1. Here, a closed space 11 shows the chamber sterilized by the approach concerning this invention, and is not restricted to the volume or configuration of the space. Specifically, medicine manufacture or the clean room for pharmaceutical preparation, the operating room of a hospital, the prosectorium, a laboratory, etc. are mentioned as this chamber.

[0011] Moreover, as for this space, being sealed substantially is desirable. That is, it is desirable in order that the bactericidal effect by this invention which that this space seals in operation explains below which carried out inside \*\* stable enough may obtain. Especially the approach of sealing this space is possible by it not being limited but sealing up clearance, such as a gate of closing and this chamber, for an indoor air regulatory system (it setting to drawing 1 and they being the pumping systems 12 and 13 of an indoor air regulatory system) etc.

[0012] In this space, the sensors 15, 16, and 17 which act as the monitor of the concentration, such as temperature in this space, humidity, and formaldehyde, are formed. the number and installation of installation of these sensors -- especially -- a limit -- there is nothing -- the volume of this space, or an internal configuration -- suitable number \*\*\*\*\* -- things are desirable.

[0013] Data processing which the data from these sensors are sent to a control system 14, and is

explained below is performed.

[0014] Moreover, in this space, the thermostat 20 is further formed with the formaldehyde generator 18 and humidity control equipment 19. the number and installation of installation of these equipments -- especially -- a limit -- there is nothing -- the volume of this space, or an internal configuration -- suitable number \*\*\*\*\* -- things are desirable. With said control device 14, the equipment to apply generates the formaldehyde of suitable concentration, and the humidification and dehumidification of it are carried out so that it may become the relative humidity of the set-up range, and it carries out heating cooling so that it may become the temperature of the range set up further.

[0015] Therefore, according to the approach concerning this invention, the temperature in this space will be maintained in the predetermined laying temperature range by the control unit 14, the thermo sensor 15, and the thermostat 20. Moreover, according to the approach concerning this invention, the humidity in this space (relative humidity to said temperature) will be maintained by the predetermined setting-out humidity requirement with a control unit 14, a humidity sensor 16, and humidity control equipment 19. Under the conditions of the starting predetermined temperature within the limits and humidity, the formaldehyde concentration in this space will be maintained in a predetermined setting range by the control device 14, the formaldehyde sensor 17, and the formaldehyde generator 18. Furthermore, there is no time limit which maintains the temperature, the specific humidity, and specific formaldehyde concentration of the range under the control condition concerning this invention.

[0016] Each equipment is explained in detail below.

[0017] (Generating of formaldehyde) It is easy to generate dew condensation by condensation of the poured moisture, and in the case of the formalin gas which contains a lot of moisture simultaneously, formaldehyde oxidizes easily, and tends to produce impurities, such as a formic acid, in it. Under the present circumstances, the formic acid contained in the dew condensation to produce may adhere to an indoor wall and an indoor vessel, and may produce the problem of contamination.

[0018] On the other hand, as for the bactericidal effect by formaldehyde, it is desirable to generate formaldehyde from correlating also with lifting of humidity being known, holding the optimal humidity.

[0019] In the approach concerning this invention, a desirable usable formaldehyde generating means is a means by which the so-called dry formaldehyde gas can be generated. That is, formaldehyde is a means only accompanied by generating of a small amount of moisture of controllable extent.

[0020] concrete -- a methanol -- from -- (-- one --) -- a catalyst -- using -- generating -- making -- a means -- (-- two --) -- sonication -- carrying out -- generating -- making -- a means -- (-- three --) -- UV irradiation -- carrying out -- generating -- making -- a means -- etc. -- mentioning -- having. Especially in this invention, (1) is desirable. Under the present circumstances, it is very little although water subgenerates simultaneously.

[0021] About the catalyst decomposition reaction of the methanol to apply, it is already known, and optimizing easily is possible about a complement and the generating conditions of the formaldehyde of purity in this invention.

[0022] Concretely, one outline of the equipment of the generating system which uses a catalyst for drawing 2 was shown. As a catalyst 21, those mixture, such as platinum, copper, aluminum, or carbon, is mentioned. The cylinder-like container 22 is filled up with the starting catalyst, and the thermostat 23 is formed in this cylindrical cup 22. Therefore, this catalyst is heated and cooled by predetermined temperature. The methanol 24 of a predetermined amount is first sent to the evaporation section 26. The evaporation section 26 evaporates a methanol with the heating cooling system 27, is sent to this catalyst part 21, and starts catalytic reaction with said thermostat 23. It is supposed that a methanol is become formaldehyde 25 and water by the reaction expressed with the following general formulas.

[0023]

$\text{CH}_3\text{OH} \longrightarrow \text{HCHO} + \text{H}_2\text{O}$  -- here, about control of the yield of formaldehyde, it is dependent on the amount of the methanol which the temperature of the catalyst by this thermostat controls and supplies, or the amount of evaporation in this evaporation section. Optimization of the starting reaction condition generates formaldehyde with actual equipment, and is possible by suitable formaldehyde density

measurement. Specifically, it is possible to create a calibration curve from the measurement data of catalytic-reaction temperature and the yield of formaldehyde gas to the predetermined methanol amount of supply. For example, when copper is used as a catalyst and it is used methanol 1000g, it is possible to generate formaldehyde gas 750g in about 30 minutes. In addition, although the heating cooling system 27 is shown by drawing 2 in order to make a methanol evaporate, it is also possible to use the evaporation approach by sonication etc.

[0024] Furthermore, as a means to generate formaldehyde, although ultrasonic irradiation processing or UV irradiation processing is possible to a methanol, when starting, it is possible by putting a methanol into a suitable reaction container and forming ultrasonic irradiation processing or a UV irradiation processor in the periphery or the interior of this reaction container (not shown).

[0025] The control system 14 shown by drawing 1 acts as the monitor of the concentration of this closed space, and when higher than the density range set up beforehand, the thermostat 23 of said generator is controlled, cooling etc. carries out a catalyst, and it controls a reaction.

[0026] Moreover, in this invention, the approach of heating a paraformaldehyde and generating as an approach of generating formaldehyde gas, is also desirable. In this case, especially a limit does not have a heating means. Moreover, the amount of generating gas becomes controllable easily by controlling whenever [ heating time / starting / and stoving temperature ]. The paraformaldehyde is usually available as a commercial item.

[0027] (Formaldehyde concentration in a closed space) In this invention, measurement of the formaldehyde concentration in a closed space can usually be based on a well-known analysis means. Specifically, a method of chemical analysis or a physical chemistry analysis method is mentioned.

Especially in this invention, it is desirable for it to be necessary to measure concentration of formaldehyde gas to ONTAIMU, and to be based on a formaldehyde sensor. Or the gas-chromatograph analysis by the air sampling in this space or ion chromatographic analysis is desirable.

[0028] According to the formaldehyde generation-of-gas means by disassembly of the above-mentioned methanol, high-concentration formaldehyde can be generated in a short time. It is possible by specifically installing the formaldehyde gas generator of the suitable number according to the volume of the space which should carry out sterilization processing, and a configuration.

[0029] The concentration of the formaldehyde gas generated in the approach concerning this invention can be easily maintained to 2000 ppm or more by using the above-mentioned generator. Furthermore, in order to acquire the bactericidal effect (or the sterilization effectiveness) explained below, it is possible to also make it generate in high concentration more. Specifically, 3000 ppm or more and 4000 more ppm or more (5000 more ppm or more) are possible.

[0030] (Temperature control means in a closed space) In this invention, in order to carry out long duration maintenance of the suitable formaldehyde gas concentration of concentration within the limits of suitable temperature, it is desirable to adjust the temperature in this closed space. There is no temperature control means established in order to start, and it is [ especially a limit ] usually usable. [ of well-known heating or a cooling system ] Under the present circumstances, although it is dependent also on the volume of space, and its configuration, it can adjust to extent which can disregard the variation in temperature substantially by using the equipment which has sufficient heat exchange capacity. In the approach concerning this invention, a desirable temperature requirement is 20 degrees C to 50 degrees C, and the range of it is 25 to 35 degrees C more preferably.

[0031] (Humidity control means in a closed space) In this invention, in order to carry out long duration maintenance of the suitable formaldehyde gas concentration of concentration within the limits of suitable humidity, it is desirable to adjust the humidity in this closed space. There is no humidity control means established in order to start, and it is [ especially a limit ] usually usable. [ of well-known humidification or an air dryer ] Under the present circumstances, although it is dependent also on the volume of space, and its configuration, it can adjust to extent which can disregard the variation in humidity substantially by using the equipment which has sufficient humidification or dehumidification capacity.

[0032] Although it is known that the bactericidal effect by formaldehyde gas will be dependent on

humidity, when it becomes high more than extent with relative humidity, dew condensation will occur according to a \*\*\*\* phenomenon, and formaldehyde or the formic acid which is the oxide will adhere to a wall etc. by condensing in this space. In this case, the inside of this space will be polluted.

[0033] Therefore, in this invention, it becomes important to maintain the humidity which is extent to which the bactericidal effect of formaldehyde is demonstrated enough and the phenomenon of above-mentioned dew condensation does not happen. Although it is dependent on temperature, as for the range of the applied humidity, it is desirable that a temperature requirement is 90% of range (from 80% to 90% [ Preferably ]) from 50% of relative humidity preferably in the range of 20 degrees C to -50 degree C. Bactericidal effect sufficient at humidity lower than the starting range is not acquired, and when higher than the starting range (90% or more), contamination may arise according to generating of dew condensation.

[0034] It is possible for especially a limit not to have the above-mentioned humidity control approach, and to usually use it combining a commercial humidifier and a dehumidifier, or them. Furthermore, also about the control approach of the equipment to apply, there is especially no limit, is manual or can be controlled automatically. It is also possible to control so that it is maintained by the humidity requirement beforehand set up according to the control system 14 shown in drawing 1 .

[0035] (Temperature monitor means) It is possible for there to be especially no limit in an approach to act as the monitor of whenever [ room air temperature / of a closed space ], and to use the usual thermometer. Moreover, in order to input into a control system 14 the temperature which acted as the monitor, an input or inputting automatically are manually possible. Therefore, in this control system 14, the temperature in this space in specific time amount will be memorized.

[0036] Although there is especially no limit also about the precision of this temperature that acts as a monitor, there should just be the about \*\*1-degree C accuracy of measurement. Furthermore, it is also possible to establish these two or more monitor means, in this case, a monitor is possible for the variation in this temperature in the interior of a room, and the more exact temperature control of it becomes possible.

[0037] (Humidity monitor means) It is possible for there to be especially no limit in an approach to act as the monitor of the indoor humidity of a closed space, and to use the usual hygrometer. Moreover, in order to input into a control system 14 the humidity which acted as the monitor, an input or inputting automatically are manually possible. Therefore, in this control system 14, the humidity in this space in specific time amount will be memorized.

[0038] Although there is especially no limit also about this accuracy of measurement that acts as a monitor, it is a 20 to 50 degrees C temperature requirement, and there should just be about \*\*1% of accuracy of measurement. Furthermore, it is also possible to establish these two or more monitor means, in this case, a monitor is possible for the variation in this humidity in the interior of a room, and the more exact humidity control of it becomes possible.

[0039] (Formaldehyde concentration monitor means) It is possible for there to be especially no limit in the approach of acting as the monitor of the indoor formaldehyde concentration of a closed space, and to use the usual analysis means. Specifically, the approach using the sensor for formaldehyde and the approach by the gas chromatograph by air sampling and the ion chromatograph are mentioned. In order to input into a control system 14 this concentration that acted as the monitor, an input or inputting automatically are manually possible. Therefore, in this control system 14, this concentration in this space in specific time amount will be memorized.

[0040] Although there is especially no limit also about this accuracy of measurement that acts as a monitor, it is the above-mentioned activity density range, and there should just be the about \*\*10 ppm accuracy of measurement. Furthermore, it is also possible to establish these two or more monitor means, in this case, a monitor is possible for the variation in this this concentration in the interior of a room, and this more exact concentration accommodation of it is attained.

[0041] (Control means) this invention -- setting -- the temperature, humidity, and formaldehyde concentration in a closed space -- the predetermined range -- it is -- predetermined time amount maintenance -- it is necessary to carry out The concentration of the formaldehyde gas which occurred in

this space decreases by various reactions, such as a sterilization reaction, in this space. Therefore, in order to maintain the concentration of formaldehyde gas uniformly, it is necessary to control a formaldehyde generating means to incorporate temperature, humidity, and formaldehyde concentration data in the setup time, and to become the specific range. Although there is especially no limit about the control approach for this object, the automatic control using the approach by hand control or a computer program is mentioned. In this invention, since it is necessary to carry out long duration maintenance of the high formaldehyde concentration, the controllable automatic-control approach is desirable in a formaldehyde generator, optimizing to ONTAIMU.

[0042] Although there is especially no limit also about the configuration of the automatic controller to apply, preferably (1) Input means, such as laying temperature, setting-out humidity, and setting-out formaldehyde concentration (keyboard etc.), (2) Temperature, humidity, a means to memorize the measurement data from a formaldehyde concentration monitor (memory etc.), (3) It has a control signal output to the output means (a screen display or printing) of those values, a means to distinguish the difference of (4) this measurement data and this set point, (5) temperature regulatory system, a humidity control system, and a formaldehyde generation-of-gas system. Here, it makes it possible to supply delivery and a raw material methanol for a control signal to a formaldehyde generator with the means of the above (5), or to raise catalytic-reaction temperature, and to make this gas yield increase with the means of the above (4), when formaldehyde concentration is lower than the set point.

[0043] (Bactericidal effect measuring method) Further, there are not a measuring method of the bactericidal effect by the approach concerning this invention and the target bacteria, either, and various well-known approaches can apply especially a limit. Specifically, the thing of an ISO standard is mentioned. In this invention, the biological indicator of commercial various configurations is usable especially with repeatability sufficient simple. The thing of a SUTORIPPUSU mold (test paper type) and the thing of a proof mold are specifically usable, and *Bacillus subtilis*, var. *niger* (ATCC No 9372), and *Bacillus stearothermophilus* (ATCC No 7953) are preferably usable as usable bacteria.

[0044] Moreover, in order to judge a bactericidal effect, after carrying out formaldehyde gas sterilization processing using the approach concerning (2) this inventions, the (1) above-mentioned test paper etc. is usually installed in the location where the plurality in a closed space was chosen, (3) this test paper is used, a suitable culture medium is cultivated, it is the existence of the survival bacteria and negative (with no survival bacteria) or a positivity (those with survival bacteria) is distinguished. Therefore, while the starting distinction approach shows a bactericidal effect, it also shows the sterilization effectiveness.

[0045] Specifically, in *Bacteria Bacillus subtilis* and var. *niger* (ATCC No 9372), the culture for seven days, and in *bacteria Bacillus stearothermophilus* (ATCC No 7953), this culture condition is preferably usable [ the culture more than for seven days ] at a TORIPU sow thicket ion culture medium (55-60 degrees C \*\*1.0 degrees C) in a TORIPU sow thicket ion culture medium (30-35 degrees C \*\*1.0 degrees C).

[0046] Specifically, the bactericidal effect by the above-mentioned distinction approach at the time of using the approach concerning this invention shows negative (that is, sterilization is meant), when the thing of *Bacillus subtilis*, var. *niger* (ATCC No 9372), and 106 order is used and the thing of *Bacillus stearothermophilus* (ATCC No 7953) and the order of 106 is used.

[0047] (The after-treatment approach) The approach concerning this invention prevents the oxidative degradation to that formaldehyde etc. adheres to the wall of a chamber etc., the formic acid of formaldehyde, etc. by preventing generating of dew condensation by controlling temperature, humidity, and formaldehyde gas concentration, and controlling especially humidity within suitable limits. Therefore, after using the approach concerning this invention, it is possible by suspending generating of formaldehyde gas and only ventilating the air in this space to end without producing contamination by the residue in this closed space.

[0048] Moreover, in case it exhausts to the open air, it is desirable for a means to perform harmless processing of exhaust gas separately, and to discharge.

[0049] How to start this invention below is explained based on an example. However, this invention is not restricted to the starting example.

[0050]

[Example] The five above-mentioned generators (formaldehyde gas 750g/hour) were installed in the cube closed space of 3 area 2 and volume of about 1000m of about 250m. Moreover, about 150 BI(s) (B. Stearo 106 and B.Subt 106) were installed in bactericidal effect measurement at every place. It acted as the monitor of the formaldehyde gas concentration by eight places (inside of a floor line, a head-lining side, and a booth), and the average value was made into formaldehyde gas concentration. Whenever [ this space internal temperature ] controlled 30 degrees C - 35 degrees C and relative humidity in 70% - 80% of range.

[0051] Spacial concentration amounted to 2000 ppm or more in about 2 hours and 30 minutes after formaldehyde generation-of-gas initiation, and it was set to 2500 ppm or more about 3 hours after. Formaldehyde concentration was maintained to about 3000 ppm after that for about 7 hours or more. Time amount change of temperature, humidity, and formaldehyde concentration was shown in drawing 3.

[0052] Indoor ventilation was carried out after suspending generating of formaldehyde gas. Then, as a result of inspecting BI, the negative result was obtained altogether 150.

[0053] Furthermore, the culture-medium restoration trial after sterilization processing (about 6000 / batch) was cleared altogether 3 times (negative decision is meant). In addition, inspection of an adhesion bacillus and a floating bacillus was cleared.

[0054]

[Effect of the Invention] According to this invention, it becomes possible to carry out long duration maintenance of the formaldehyde gas stably [ are high concentration and ] at the optimal humidity in a closed space. Therefore, repeatability is good and sufficient bactericidal effect is demonstrated. Furthermore, according to the approach of this invention requiring, since the optimal temperature and humidity can be adjusted, in this closed space, the contamination residues, such as a paraformaldehyde and a formic acid, do not exist substantially after this sterilization processing.

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## DESCRIPTION OF DRAWINGS

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### [Brief Description of the Drawings]

[Drawing 1] Drawing 1 is drawing having shown the approach concerning this invention typically.

[Drawing 2] Drawing 2 is drawing having shown the outline of the formaldehyde generator used by the approach concerning this invention.

[Drawing 3] Drawing 3 is drawing which was measured at 20-50 degrees C and in which having shown humidity and formaldehyde concentration in the example of this invention.

### [Description of Notations]

11 -- closed space, 12, 13 -- pumping tubing, and 14 -- a control device, 15 -- temperature monitor, 16 -- humidity monitor, and 17 -- a formaldehyde concentration monitor, 18 -- formaldehyde generator, 19 -- humidification-and-dehumidification equipment, and 120 -- a heating cooling system, 121 -- after-treatment equipment, 21 -- catalyst, and 22 -- a cylindrical cup 23 -- thermostat, 24 -- methanol supply system, and 25 -- formaldehyde, 26 -- evaporation equipment, and the heating cooling system for 27 -- evaporation

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[Translation done.]







